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CIRRIS 1A OBSERVATION OF $^{13}\text{C}^{16}\text{O}$ AND $^{12}\text{C}^{18}\text{O}$ FUNDAMENTAL BAND RADIANCE IN THE UPPER ATMOSPHERE

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Abstract. Fundamental band emission from the $\nu=1$ levels of isotopically substituted $^{13}\text{C}^{16}\text{O}$ and $^{12}\text{C}^{18}\text{O}$ has been observed for the first time in the earth's atmosphere. High-resolution earthlimb spectra were obtained in the Cryogenic Infrared Radiance Instrumentation for Shuttle (CIRRIS 1A) experiment under both nighttime and daytime conditions. Spectral data between 70 and 150 km tangent height were analyzed in the 4.7 μm wavelength region, providing absolute $^{12}\text{C}^{16}\text{O}$, $^{13}\text{C}^{16}\text{O}$, and $^{12}\text{C}^{18}\text{O}$ column radiances. The minor isotope column radiances are up to 30 times more intense than predicted, based solely on the natural isotopic abundances of ^{13}C and ^{18}O . The derived line-of-sight radiances were accurately modeled using the Phillips Laboratory Atmospheric Radiance Code, and provide a strong test of radiative transport and excitation algorithms, as well as model climatologies.

Introduction

Low concentration species are known to play a dominant role in upper atmospheric infrared emission and radiative cooling. If a molecular emission band becomes optically thick, otherwise secondary features such as hot bands, satellite bands, and emissions from isotopically substituted species may contribute a greater proportion of the total intensity. In the case of the $\text{CO}_2(\nu_3)$ band at 4.3 μm , for instance, emissions from the isotopically substituted species—including $^{13}\text{CO}_2$ (1.1% terrestrial abundance), ^{18}OCO (0.4%), and ^{17}OCO (0.07%)—are magnified out of proportion to the emitter number densities. Assuming a nighttime limb line-of-sight viewing path with a 70 km tangent height, the Strategic High-Altitude Radiance Code (SHARC) [Sharma *et al.*, 1989] predicts that the isotopically substituted CO_2 species contribute fully two-thirds of the total observed radiance in the $\text{CO}_2(\nu_3)$ band, despite accounting for less than 2% of the ground-state population. This prediction has been corroborated through analysis of spectral data from the SPIRE rocketborne experiment [Wintersteiner *et al.*, 1992], and from the CIRRIS 1A space shuttle mission [Winick *et al.*, 1992].

We have previously reported a preliminary analysis of high-altitude earthlimb emission spectra from the CIRRIS 1A experiment in the 4.7 μm region [Dodd *et al.*, 1993]. Subtraction of modeled O_3 , OH, and $^{12}\text{C}^{16}\text{O}$ (main isotope) radiance leaves a series of lines spaced 3.4 cm^{-1} apart in the

2060-2150 cm^{-1} range. These lines have since been identified as a superposition of 1-0 fundamental vibration-rotation emissions in $^{13}\text{C}^{16}\text{O}$ and $^{12}\text{C}^{18}\text{O}$, and represent the first atmospheric observation of infrared emission from these species. Isotopically substituted $^{13}\text{C}^{16}\text{O}$ (1.11% of total CO) and $^{12}\text{C}^{18}\text{O}$ (0.204%) are observed to radiate with up to 30 times more intensity than expected, based solely on the isotopic abundances. These enhancements arise from radiative excitation and transport effects in the atmosphere, in analogy to the case of CO_2 .

A second possible contributor to the enhanced CO minor isotope fundamental band radiance is that of ^{13}C and ^{18}O isotope fractionation in ground-state CO. However, this hypothesis is not consistent with existing field data. Isotope partitioning in tropospheric CO has been examined through *in situ* measurements, revealing a small and variable amount (<5%) of ^{13}C and ^{18}O isotope fractionation [Kaye, 1987], *i.e.* far less than the radiative enhancements observed in the CIRRIS 1A experiment. Isotope partitioning in mesospheric CO has been investigated *via* the $\nu=0$, $J=2 \rightarrow 1$ microwave emission line using ground-based detection (R.T. Clancy, unpublished data, 1992). These emissions are consistent with a $^{12}\text{C}^{16}\text{O}/^{13}\text{C}^{16}\text{O}$ abundance ratio equal to the nominal terrestrial value of 89.

In the following, we describe the analysis of the CO spectral data, and also the use of the Phillips Laboratory Atmospheric Radiance Code (ARC) [Wintersteiner *et al.*, 1992] in generating radiance predictions. These predictions are sensitive to several model inputs and assumptions, including the local climatology, earthshine and solar flux source terms, and radiative excitation and transport algorithms. Observation of infrared radiance from the $\nu=1$ levels of isotopically substituted CO thus provides an important new test for the validation of existing models.

Experiment and spectral data analysis

Spectral data were obtained by the CIRRIS 1A experiment [Ahmadjian *et al.*, 1990], flown aboard the space shuttle Discovery during 28-30 April 1991. A cryogenic Michelson interferometer with 0.6 cm^{-1} unapodized FWHM linewidth was used to observe earthlimb infrared emission. A series of optical filters were used to isolate specific portions of the spectrum in the 400-4000 cm^{-1} range. The scans analyzed herein were obtained using the largest, most sensitive detector (Detector 2), and a 4.9 μm short-pass filter (Filter 1). A total of 26 nighttime and 66 daytime scans between 70 and 150 km tangent height were analyzed in this study. The raw interferograms were triangularly apodized prior to fast Fourier transformation. This apodization was chosen in order to diminish sidelobe intensities, and also to produce an analytic lineshape function ($\text{sinc}^2 x$), which facilitated the spectral

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fitting computations. The apodized FWHM linewidth was approximately equal to 0.95 cm^{-1} .

Figure 1 shows nighttime spectral data in the $2060\text{--}2220\text{ cm}^{-1}$ region for a tangent height of 85 km, the spectrum obtained as the average of three spectra with similar tangent heights and emission intensities. Spectra such as that shown in Figure 1 were fit using methods described in a previous publication [Dodd et al., 1993]. Observed radiance was found to be closely reproduced at all tangent heights by including six molecular bands: the O_3 $\nu_1+\nu_3$ (band origin 2111 cm^{-1}) and $\nu_1+\nu_2+\nu_3-\nu_2$ (2084 cm^{-1}) combination bands, the OH 9-8 band (2236 cm^{-1}), and the 1-0 bands of $^{12}\text{C}^{16}\text{O}$ (2143 cm^{-1}), $^{13}\text{C}^{16}\text{O}$ (2096 cm^{-1}), and $^{12}\text{C}^{18}\text{O}$ (2092 cm^{-1}). No evidence was found for the $^{12}\text{C}^{17}\text{O}$ 1-0 band (2116 cm^{-1}), consistent with the low (0.037%) $^{12}\text{C}^{17}\text{O}$ abundance. All line positions and intensities were taken from the HITRAN database [Rothman et al., 1992]. Normalized spectral profiles for the O_3 , OH, and CO minor isotope species were determined assuming a Boltzmann distribution over the rotational levels of the upper vibrational state. An effective rotational temperature T was employed as a non-linear, adjustable parameter in the spectral fits, with a single value of T applying to O_3 , OH, and the CO minor isotopes. Values of T consistent with the expected local kinetic temperature were obtained.

Unlike the other features, $^{12}\text{C}^{16}\text{O}$ main isotope emission is optically thick along the line-of-sight for the lower tangent heights. For this reason, the normalized $^{12}\text{C}^{16}\text{O}$ 1-0 spectral profile was determined from a SHARC calculation, for conditions appropriate to the scan being fit. Each of the synthetic spectral basis functions, thus generated, was scaled independently in the spectral fitting process, with the fit determining the optimum multipliers in order to minimize the sum of the squares of the residuals (χ^2). The $^{13}\text{C}^{16}\text{O}$ and

$^{12}\text{C}^{18}\text{O}$ basis function multipliers were found to be only slightly correlated ($|r| = 0.2\text{--}0.4$); both were uncorrelated with the $^{12}\text{C}^{16}\text{O}$ basis function multiplier.

The bottom of Figure 1 shows the spectral fit corresponding to the scan at the top. The sequence of nearly equally spaced lines between 2145 and 2220 cm^{-1} results from rotational transitions in the R -branch of the $^{12}\text{C}^{16}\text{O}$ 1-0 band. At lower wavenumber values, sharp lines due to the three CO isotopic species are seen superimposed on the broader O_3 emission. Figure 2 shows the fitted contributions made by the three CO isotopic species. In general, the CO minor isotope emissions are weak, and can only be detected through subtraction of overlying emission which has been characterized through spectral fitting techniques. Because the filter transmittance decreases by a factor of ten between 2100 and 2060 cm^{-1} , the CO 1-0 P -branches are observed with much lower S/N than the R -branches. However, since the residuals are weighted according to the calibration function, discrepancies between the data and model intensities below 2100 cm^{-1} contribute little to the overall value of χ^2 .

Finally, for the purposes of generating integrated column radiances, the model CO 1-0 vibration-rotation line intensities were summed, and the sum multiplied by the appropriate best-fit column density from the spectral fit. Similar integrated radiances were determined from fits to selected spectra generated from unapodized data, indicating that the calculated radiances do not depend on the choice of the apodization function.

Model radiance calculations

The ARC model has been used to perform detailed calculations of non-LTE excitation and radiance for the $^{12}\text{C}^{16}\text{O}$ main isotope [Winick et al., 1993]. In this work, earthlimb radiance from all three CO isotopes was computed

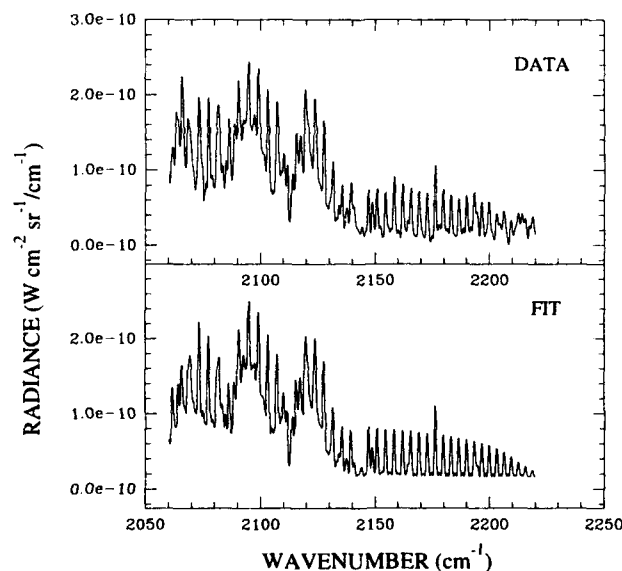


Fig. 1. Nighttime spectral data at 85 km tangent altitude (top) and corresponding spectral fit (bottom), with the data obtained as the average of CIRRIS 1A scans 11b 0677, 11b 0678, and 12b 0546. Observed radiance is from the O_3 $\nu_1+\nu_3$ and $\nu_1+\nu_2+\nu_3-\nu_2$ combination bands, and from the OH 9-8 and CO 1-0 fundamental bands.

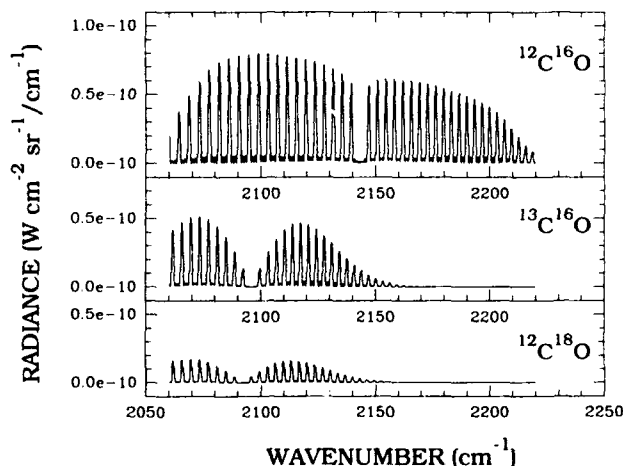


Fig. 2. Synthetic spectral basis functions for the 1-0 fundamental bands of $^{12}\text{C}^{16}\text{O}$ (top), $^{13}\text{C}^{16}\text{O}$ (middle), and $^{12}\text{C}^{18}\text{O}$ (bottom), scaled according to their contributions to the spectral fit function shown in Figure 1. The $^{12}\text{C}^{16}\text{O}$ profile was obtained from the SHARC line-of-sight model [Sharma et al., 1989], while the minor isotope profiles were generated assuming negligible self-absorption and the best-fit rotational temperature (193 K).

for the 70–130 km tangent altitude range. Spectral line positions and intensities for all species were taken from the 1992 HITRAN database [Rothman et al., 1992]. Reaction and collisional energy transfer rate coefficients for the minor isotopes were taken to be identical to those used for $^{12}\text{C}^{16}\text{O}$. Isotopic abundances were assumed to be independent of altitude in the range under study. The kinetic temperature and N_2 , O_2 , and O densities were taken from standard climatology models [Summers, 1993; Hedin, 1991], assuming the month of April and 40°S latitude, together with the "medium" CO profile from Lopez-Puertas et al. [1993]. The CO mixing ratio was assumed to be proportional to the N_2 mixing ratio for altitudes above 120 km. Using a similar climatology, a set of 100 CIRRIS 1A $^{12}\text{C}^{16}\text{O}$ earthlimb radiance values in the 70–140 km tangent height range were satisfactorily predicted by ARC, to within a factor of two in all cases [Winick et al., 1992]. Previous calculations have investigated the sensitivity of the CO 1–0 limb radiance to the kinetic temperature and CO concentration profiles [Winick et al., 1993; Lopez-Puertas et al., 1993].

Figure 3 shows $^{12}\text{C}^{16}\text{O}$, $^{13}\text{C}^{16}\text{O}$, and $^{12}\text{C}^{18}\text{O}$ earthlimb radiances derived from the spectral fits, under nighttime (top) and daytime (bottom) conditions, together with predictions made by the ARC model. Derived intensities less than about $7 \times 10^{-11} \text{ W cm}^{-2} \text{ sr}^{-1}$ cannot be distinguished from the noise in the spectra. In the nighttime data, all of the CO limb radiances lie within a factor of ten of each other at a given tangent height, despite the overwhelming prevalence of the $^{12}\text{C}^{16}\text{O}$ main isotope. The large contribution made by the minor isotopes to the total CO radiant intensity is due to two factors. First, self-absorption reduces the line-of-sight $^{12}\text{C}^{16}\text{O}$

main isotope intensity by as much as a factor of two at the lower tangent altitudes. Second, and more significant, earthshine pumping of the $\nu=1$ level is much greater for the isotopically substituted species than for $^{12}\text{C}^{16}\text{O}$ [Winick et al., 1993]. Upper atmospheric $^{13}\text{C}^{16}\text{O}$ and $^{12}\text{C}^{18}\text{O}$ are efficiently radiatively excited by the warm lower atmosphere and ground boundary, while the optically thick $^{12}\text{C}^{16}\text{O}$ is largely pumped by cooler CO at higher altitudes. ARC predictions for the daytime and nighttime vibrational temperatures (T_{vib}) for the CO isotopic species are shown in Figure 4. Above 60 km altitude, the nighttime minor isotope T_{vib} values are predicted to be 40 K higher than $T_{\text{vib}}(^{12}\text{C}^{16}\text{O})$, corresponding to a factor of ten enhancement in the minor isotope $\nu=1$ populations.

During the daytime, the minor isotope contribution to the total CO radiance is smaller (Figure 3), since pumping of $\nu=1$ in all of the isotopes arises largely through solar excitation. Solar radiation also raises the CO vibrational temperature in the middle atmosphere, thus increasing the source function for pumping of $^{12}\text{C}^{16}\text{O}$ $\nu=1$. Nevertheless, the minor isotope T_{vib} values are still greater than $T_{\text{vib}}(^{12}\text{C}^{16}\text{O})$, since the effective earthshine source temperature for pumping of the minor isotopes (288 K) is significantly higher than that for pumping of $^{12}\text{C}^{16}\text{O}$ (260 K). In addition, the solar radiant intensity is attenuated by as much as 30% on the $^{12}\text{C}^{16}\text{O}$ lines, owing to absorption by CO in the sun's atmosphere [Farmer and Norton, 1989]. The predicted 16 K difference in T_{vib} , shown in Figure 4, corresponds to a 2.5-fold enhancement in the minor isotope $\nu=1$ populations.

In Figure 3, the daytime $^{13}\text{C}^{16}\text{O}$ and $^{12}\text{C}^{18}\text{O}$ radiances above 80 km were obtained from spectral data averaged within three different 5 km tangent height bins. Fits performed to single scans in this range displayed a factor-of-three degree of scatter in the radiance values. The daytime spectra were obtained over a wide variety of locations on the earth's surface (6°S to 70°N latitude). Thus, the scatter may

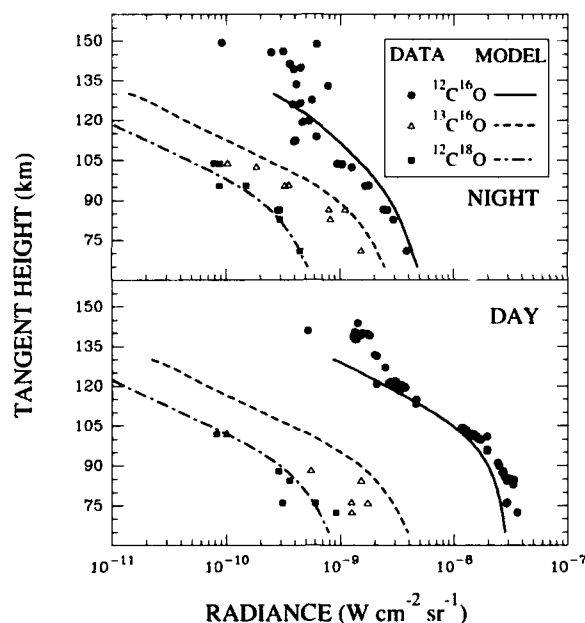


Fig. 3. Integrated radiance from the 1–0 fundamental bands of $^{12}\text{C}^{16}\text{O}$, $^{13}\text{C}^{16}\text{O}$, and $^{12}\text{C}^{18}\text{O}$, as determined from spectral fits to numerous scans from the CIRRIS 1A database, under nighttime (top) and daytime (bottom) conditions. The lines superimposed on the data points are predictions made by the Atmospheric Radiance Code (ARC) model [Wintersteiner et al., 1992].

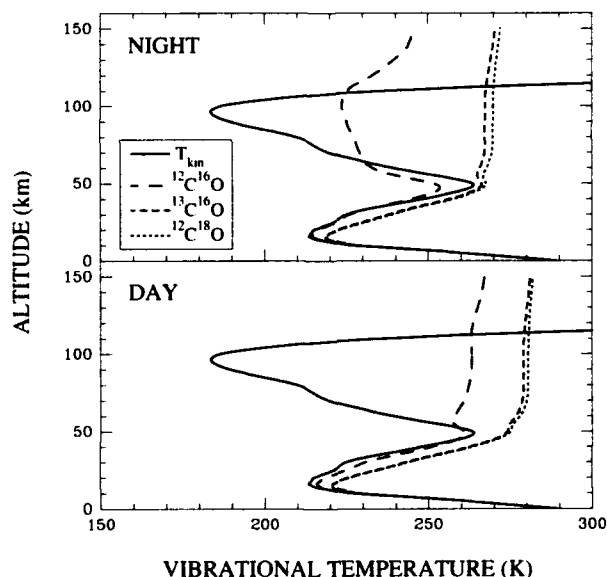


Fig. 4. Nighttime (top) and daytime (bottom) vibrational temperature profiles for $^{12}\text{C}^{16}\text{O}$, $^{13}\text{C}^{16}\text{O}$, and $^{12}\text{C}^{18}\text{O}$, as predicted by the ARC model. Also shown for reference is the kinetic temperature input to the ARC calculation.

result from variations in the local cloud cover and surface characteristics, both of which strongly influence radiant pumping from the lower boundary. However, no obvious correlations were observed between the derived CO minor isotope radiance and geographical location, partly due to the relatively small amount of data. Instead, we attribute the scatter to the difficulties inherent in measuring the weak CO minor isotope emission in the presence of the factor of 20-200 stronger $^{12}\text{C}^{16}\text{O}$ main isotope radiance. The scatter is less evident in the nighttime data, in which the minor isotopes contribute a much greater fraction of the total radiance.

For both night and day, the model predicts increasing $^{12}\text{C}^{16}\text{O}$ self-absorption at lower tangent altitudes, which in turn increases the minor isotope contribution; this prediction is in accord with the data. For tangent heights above about 120 km, the ARC model significantly underpredicts the $^{12}\text{C}^{16}\text{O}$ radiance, the discrepancy increasing with tangent height. This discrepancy may arise from an underestimate of the CO mixing ratio for those altitudes, and will be investigated in future work.

Summary

Detailed spectral fitting techniques, applied to unique limb radiance data, have enabled the first remote detection of $^{13}\text{C}^{16}\text{O}$ and $^{12}\text{C}^{18}\text{O}$ infrared radiance in the earth's upper atmosphere. Vibration-rotation emission lines in the 1-0 fundamental bands of $^{12}\text{C}^{16}\text{O}$, $^{13}\text{C}^{16}\text{O}$, and $^{12}\text{C}^{18}\text{O}$ were analyzed in the 70-150 km tangent height range, under quiescent nighttime and daytime conditions. The CO minor isotope radiant intensity is far greater than predicted based solely on the relative isotopic abundances, especially during the night and at low tangent altitudes. The ARC line-by-line radiative excitation model was used to reproduce the experimental observations. CO minor isotope radiance observed concurrently with $^{12}\text{C}^{16}\text{O}$ radiance provides an important test of models of radiative excitation and transport, as well as model climatologies.

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